Characterization of the high-energy excitations in two-dimensional, topological Bi_2Se_3: unusual surface transitions and cooling dynamics

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Macroscopic Bi_2Se_3 is a layered material consisting of a high number (>10) Se/Bi/Se/Bi/Se quintuple layers, separated by van der Waals gaps. The material has been characterized as a topological quantum spin Hall insulator, with an inverted bandgap of 300 meV, protecting helical surface states. Regarding the optical properties, the absorption spectrum shows intriguing optical transitions in the 0.5 – 3.5 eV energy region: for instance, a strong optical transition at 2.8 eV located at the surface, followed by luminescence in the region of 2.3 eV, preserving chirality (1,2).

The question is what happens if the system is brought into the two-dimensional regime, i.e., with a reduced number of QLs? We used colloidal two-dimensional Bi_2Se_3 nanoplatelets with a thickness of 6 QLs as a model system and studied the optical transitions in the 1-3 eV region with absorption and transient absorption quenching spectroscopy (3). We observe more than 10 strong optical transitions. Comparison with advanced DFT-GW theory allowed us to identify these transitions, group them as surface and interior transitions, and locate them in the two-dimensional Brillouin zone.
Example: When pumped in the 2.6 – 2.9 eV region - mostly a Rashba Shaped Surface band (RSS) – topological surface band transition (SS2) - we observe a constant absorption quenching in the time region below 10 ps, followed by mono-exponential decay of the quench in the time region 10 ps – 1 ns. This unusual kinetics is understood to be due to the initial separation of electrons (in the SS2 band) and holes (in the deformed and valleyed RSS band) in momentum space upon initial cooling, preventing recombination. After 10 ps, the holes gradually cool towards the Γ point by RSS → valence band transfer, where they recombine with the majority carriers, explaining the common recombination kinetics (single exponential) that we observe in the 10 ps – 1 ns time region. With the advanced DFT-GW model, it is also possible to understand the results of Ref.1, i.e. the strong "surface state transition", followed by chiral luminescence of electron in the SS2 band and holes in the valley of the RSS band (on the Γ-K line). Two-dimensional Bi_2Se_3 does not only feature as an electronic topological insulator, but also as a remarkable opto-electronic material!

1. Observation of chiral surface excitons in a topological insulator Bi2Se_3. Proceedings of the National Academy of Sciences of the United States of America 116, 4006-4011 (2021)
2. Complex optical conductivity of Bi2Se3 thin film: Approaching two-dimensional limit Applied Physics Letters 118, DOI: 10.1063/5.0049170 (2021)
3. Identification of high-energy excitations in two-dimensional topological Bi2Se3 platelets. Manuscript in preparation